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Energy and Technology Review

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LAWRENCE LIVERMORE LABORATORY ENERGY AND TECHNOLOGY REVIEW

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June 1975

Briefs

The short items on this page announce recent developments. Some of these items may be amplified in future issues. None of this material is reported elsewhere in this issue; summaries of this month's articles appear on the opposite page.

URANIUM-235 FISSION CROSS SECTION ACCURATELY DETERMINED

We have measured the fission cross section of uranium-235 relative to the standard reaction (neutron-proton scattering) for incident neutron energies between 0.8 and 20 MeV. The goal of 1 to 2% accuracy for the $^{235}U(n,f)$ cross section was achieved in the 0.8- to 14-MeV energy region. Work is continuing to extend the data down to 1 keV and to normalize them to thermal fission cross sections.

The accuracy of these measurements was made possible by our flux-monitoring system and by the unprecedented use at these energies of the time-of-flight method with a Linac source. Because of this accuracy, the $^{235}U(n,f)$ reaction may now be used as a flux standard for other fission cross-section measurements. These new data, published in the *Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D.C., March 3-7, 1975*, are more accurate than previous work.

SEARCH FOR ANTINEUTRINO DECAYS PROVES FRUITLESS

It was discovered recently that the flux of neutrinos reaching the earth from the sun is much less than predicted by present theories. One hypothesis has been that the neutrinos decay before reaching the earth. From symmetry arguments, if neutrinos decay, so should antineutrinos; thus some evidence of antineutrino decay might be found in the intense fluxes of antineutrinos generated in the explosion of nuclear fission devices. Using a large-volume sodium-iodide detector, we fielded antineutrino-decay experiments in three nuclear test events. No such evidence was found.

Complicating the experiments was the fact that present theories of particles subject to weak interactions do not predict neutrino decay and, hence, offer no help in suggesting possible decay products. Our approach was to look for any type of decay process that proceeds with the emission of products having kinetic energies up to that of the antineutrinos themselves (presumably about 0.5 MeV). This included processes as pair production.

We attempted to measure the of the incident antineutrino flux that appears .e kinetic energy of decay particles. No such particles were observed. Thus we find no evidence to support the possibility that antineutrinos decay by means of any products that can deposit 2 to 500 keV of energy in a sodium-iodide detector.

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As part of the CTR research program, we are investigating the feasibility of the gas-embedded Z-pinch, a new version of an old thermonuclear fusion concept. The Z-pinch is attractive because it is basically simple and could be adapted to a liquid-lithium-wall reactor design, bypassing first-wall 14-MeV neutron damage problems. Its chief drawback is the short growth time of instabilities. Recent theoretical and computational results have been encouraging, however, and preliminary experiments are now under way.

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We are working on an in situ *process for recovering oil from thick oil-shale deposits of moderately low grade. The concept, called rubble* in situ *extraction (RISE), consists of breaking up the oil shale into rubble underground by mining methods, then retorting the rubble in place. We estimate that this process could be developed in about 6 years at a cost of about $80 million with a high probability of success. Successful development would make it possible to produce shale oil at lower economic and environmental costs than conventional surface retorting and would add significantly to our domestic oil supply.*

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Our rubble in situ *extraction (RISE) concept is supported by experimental work begun two years ago as part of our nuclear rubblization program for oil-shale recovery. Thus far we have developed a kinetic model of kerogen decomposition and a computational model of the retorting process and have studied the heating of oil-shale blocks in air and nitrogen.*

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As part of an effort to understand the uptake of pollutants by marine organisms, we have monitored the accumulation of radioactive trace metals by oysters following low-level radioactive waste releases from the Humboldt Bay (California) nuclear power plant. Our results indicate that suspended particulates play an important role in the accumulation of some radionuclides and that resuspension of particulates from sediments is an important source of radionuclides between periods of releases.

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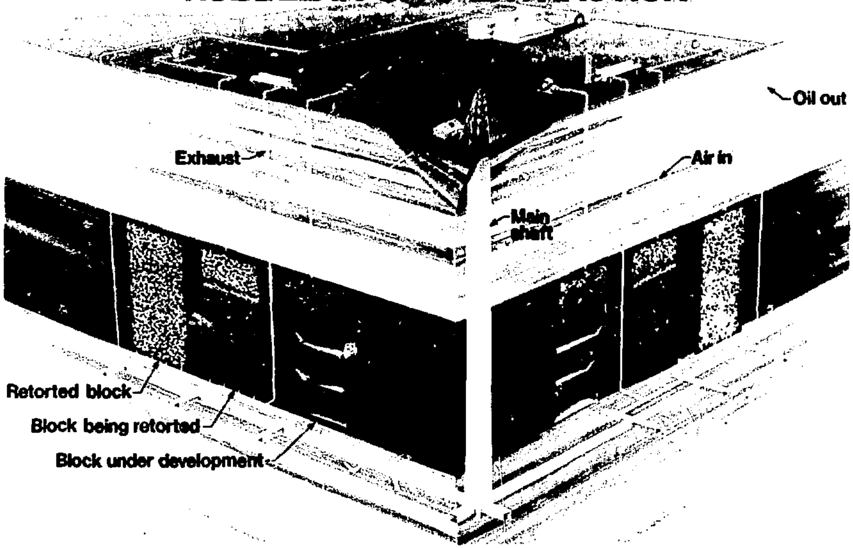
RISE
RUBBLE IN SITU EXTRACTION



Rubble *in situ* extraction (RISE) concept proposed by LLL for oil recovery from thick oil-shale deposits of moderately low grade. The RISE method, whose development appears to be relatively inexpensive, could supply oil at acceptable economic and environmental costs (see article beginning on p. 6). The process involves breaking up the oil shale into rubble underground by mining methods, then retorting the rubble in place. In Colorado's Piceance Basin alone, deposits suitable for RISE processing contain 50 × 10^9 tonnes (350 billion barrels) of recoverable oil, more than the entire Middle East oil reserves.

ADVANCED ENERGY SYSTEMS

GAS-EMBEDDED Z-PINCH: NEW APPROACH TO AN OLD FUSION CONCEPT

As part of the CTR program's exploratory plasma research, we have been investigating the feasibility of the gas-embedded Z-pinch, a high-density-plasma version of an old thermonuclear fusion concept. The Z-pinch is attractive for a fusion reactor because it is basically simple and could be adapted to a liquid-lithium-wall reactor design, bypassing first-wall 14-MeV neutron damage problems. Its chief drawback is the short growth time of instabilities, which thus far have been hard to understand and control. However, recent theoretical and computational studies have been encouraging. We are now proceeding with preliminary experiments.

Conceptually, the Z-pinch is simply a straight, high-current discharge in deuterium or D-T gas. The self-induced magnetic force constricts (pinches) the current-carrying plasma to a small radius, as shown in Fig. 1. The plasma is heated both by this pinch compression and by resistive heating by the current. Large pulsed currents of more than one million amperes are required to reach fusion temperatures of greater than ten million kelvins. The discharge is called the Z-pinch to distinguish it from another fusion concept, the theta-pinch, where the current flows around the axis rather than parallel to it.

Contact James W. Shearer (Ext. 7311) for further information on this article.

Early Z-Pinch Research at LLL

The detection of neutrons from early Z-pinch experiments stimulated considerable CTR research during the middle 1950's (Project Sherwood). Initially, neutrons were thought to arise from a single-temperature hot thermonuclear plasma. It was found, however, that they were actually produced by a small number of high-energy deuterium ions accelerated by strong, local electric fields created by "sausage" and "kink" instabilities[1] (see Fig. 2). Theoretical and experimental studies of these instabilities showed that they grow rapidly to such a magnitude that they break up the discharge and/or drive it to the wall.

The calculated growth time of the instabilities was discouraging. To exceed fusion breakeven, when the generated thermonuclear reaction energy exceeds the initial heating energy, the Lawson number (density × confinement time) must be above 10^{14} s/cm^3. At the low densities studied in the mid-1950's, the growth time for both the sausage and the kink instabilities

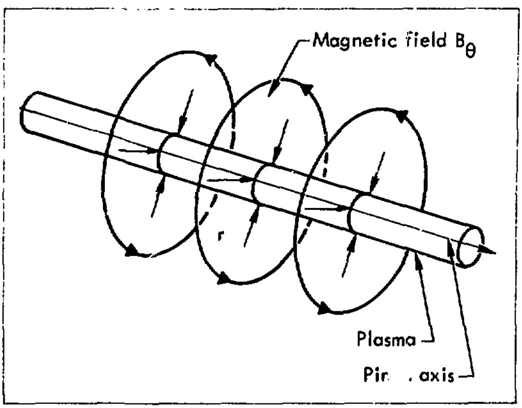


Fig. 1. Basic structure of the Z-pinch. The longitudinal electrical current flows parallel to the z-axis of the cylindrical coordinate system, inducing an azimuthal magnetic field, B_θ, whose field lines encircle the current. The force exerted on the current-carrying plasma ions by this magnetic field is directed inward, along the radial coordinate r, which constricts the current channel to a small radius.

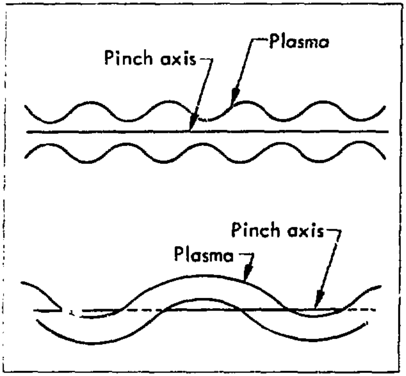


Fig. 2. Principal Z-pinch instabilities: sausage (top) and kink (bottom). When the geometry of the straight pinch is perturbed, the current and magnetic-field distributions are also perturbed in magnitude and direction. This leads to perturbations in the pinch's pattern of forces, which increase the original perturbation of pinch geometry. Thus, the amplitude of the instability continues to grow in time.

was about 1000 times shorter than the confinement time needed for appreciable fusion reactions. As a result, this early Z-pinch work was abandoned.

Recent Calculations

Recent studies at LLL have indicated that the Z-pinch instabilities may not grow as rapidly as previously believed. The sausage instability was examined on a numerical magnetohydrodynamics code[2] that calculated plasma motions in both the radial and axial directions. These calculations showed that the sausage instability leads to velocity swirls (see Fig. 3), which turbulently decay into a radial density distribution that is stable against further sausage perturbations. Thus, if the plasma is initially unstable to the sausage mode, it "heals itself" and assumes a stable configuration.

Numerical studies of the kink instability require a three-dimensional code calculation, which is only now being developed. However, two concepts that may limit or control kink instability are already partly understood. First, raising the density of the pinch reduces the ratio of instability growth time to confinement time. Calculations imply a small-radius pinch with high values of the local magnetic field in the range 10^3 to 10^4 T. Second, the gas-embedded Z-pinch (first suggested by H. Alfvén[3]) replaces the insulating wall of the conventional pinch tube with a mass of high-density, low-temperature, deuterium gas. This high-density gas constrains the growth of the kink instability without introducing wall impurities.

Gas-Embedded Z-Pinch

To produce a gas-embedded Z-pinch, we start with a high-pressure, deuterium gas fill rather than the low-pressure fill used in the earlier Z-pinch work. At present, the discharge is initiated with a relativistic electron beam; however, we are also considering other preionization methods, such as lasers or energetic ion beams. After the discharge channel is ionized and heated, the current must rise very rapidly to form the pinch. A fast capacitor-bank discharge or similar power supply providing megampere current levels is postulated.

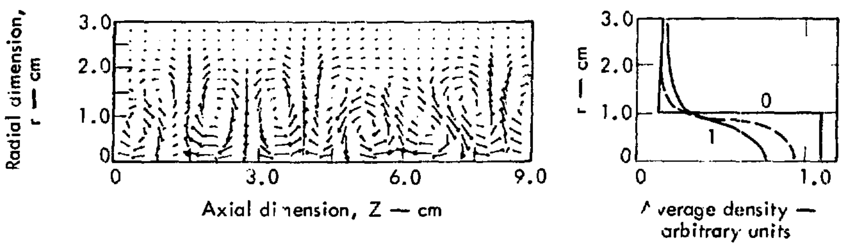


Fig. 3. Computer simulation (left) and plot of plasma density distribution vs radius (right) for the sausage instability. Curve (0) represents the chosen initial density distribution. The dashed curve is an intermediate density distribution calculated by the code after the sausage instability is fully grown. The corresponding velocity distribution is shown in the computer simulation – arrow length is proportional to velocity. At later times in the calculation, the velocities slow down and the density distribution approaches curve (1), which has been mathematically shown to be stable against further sausage perturbations.

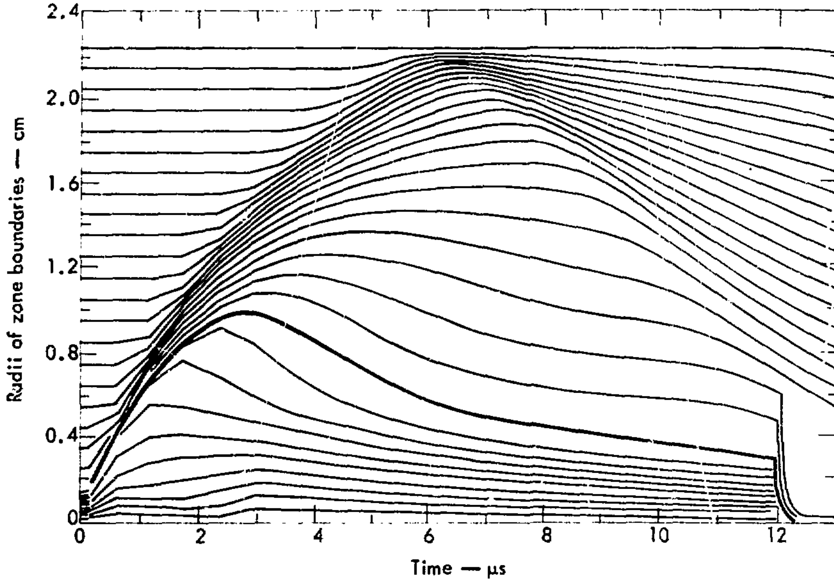


Fig. 4. Computer-calculated radius/time history of the formation of a gas-embedded Z-pinch from an initially uniform-density gas fill.

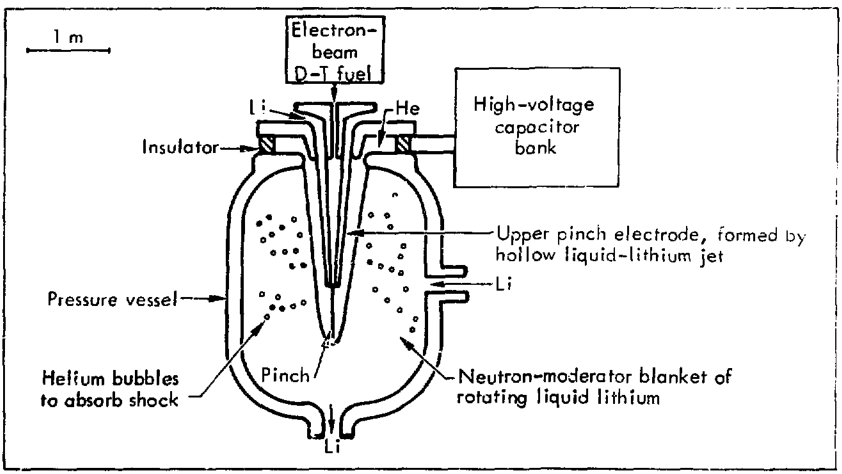


Fig. 5. Blast-containment reactor design using a Z-pinch rather than a laser-driven pellet. The paraboloidal cavity is formed by the rotation of the liquid-lithium blanket; the cavity bottom is also the lower pinch electrode.

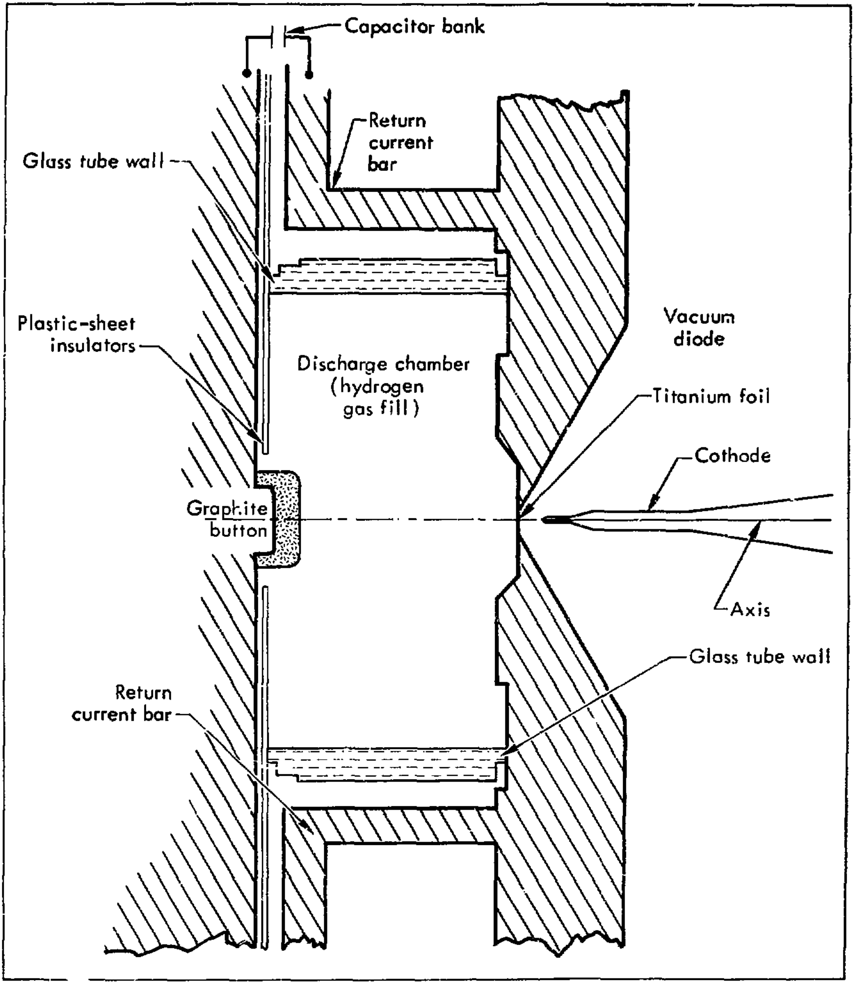


Fig. 6. Apparatus for the gas-embedded Z-pinch experiment. The vacuum diode is charged by a triggered Van de Graaf machine. The electrons are produced by field emission at the tip of the cathode and accelerated across a 5-mm vacuum gap toward a 0.025-mm-thick titanium-foil anode, thin enough to let electrons pass through but thick enough to contain the discharge chamber's 100- to 200-kPa hydrogen gas fill. The parameters of the electron-beam pulse are approximately 1.5 MeV, 20 kA, and 30 ns. A small 20-kV, 10-μF capacitor bank is directly connected across the discharge chamber, and the discharge is triggered by the electron beam. The anode of the discharge is a graphite button, its cathode the titanium foil; the axial length of the discharge is 40 mm. There are 18 return current bars separated by viewing windows for diagnostics, and the low-inductance, flat-plate line connecting the capacitor bank to the discharge chamber is insulated with plastic sheets.

Figure 4 shows a one-dimensional computer calculation for the formation of a gas-embedded Z-pinch channel. Because it is a Lagrangian calculation, the gas is radially divided into zones at time zero. At the start of the computer run, the innermost 10 zones are assumed to have been ionized by the electron-beam preionizer. For the first 3 μs, the channel expands rapidly because of ohmic heating by a 10^5-A current. Then there follows a gradual constriction as the pinch approaches equilibrium after the overshoot of the initial expansion. At 12 μs, a large pinch current of 20 MA is turned on, and the innermost 12 zones form the Z-pinch. The additional two zones are heated by thermal conduction from the initially ionized 10 zones. All other zones remain cold, forming the gas-embedding of the Z-pinch.

Because a high-density, thermonuclear Z-pinch implies high magnetic fields and explosive energy releases, design characteristics of the ultimate Z-pinch reactor must be considered from the start. One design possibility is based on the blast-containment concept first proposed for laser fusion.[4] As shown in Fig. 5, the electron beam and D-T fuel are introduced through a hollow electrode formed by a jet of liquid lithium. The other electrode is a frothy mass of liquid-lithium moderator, which is rapidly rotated to create a parabolically shaped Z-pinch discharge cavity. Such a reactor has the advantage that there is no solid wall material near the 10^3- to 10^4-T magnetic fields and high-intensity 14-MeV neutron flux. The liquid lithium contains the large magnetic pressure and absorbs the damaging radiation.

Our present experimental program is directed toward producing a gas-embedded Z-pinch, using available hardware. The apparatus is shown and briefly explained in Fig. 6.

A relativistic electron beam preionizes the gas along the axis to a diameter of about 5 mm. The characteristics of this preionization were studied in preliminary experiments using only the electron beam. We saw discharge channels of enhanced luminosity that we believe originate from space charges momentarily created by the electron beam. Because of this complicated ionization pattern, the voltage breakdown path in the high-density hydrogen gas fill is crooked. However, the mass per unit length of gas that is heated should be small, because the hottest and most highly ionized gas has the lowest resistivity. Thus, despite the crooked breakdown path, we expect that a Z-pinch will be formed that is suitable for our first experiments.

When both the electron-beam accelerator and the capacitor bank are charged, the relativistic beam breakdown path acts as the switch for the capacitor bank. The risetime to reach full current is about 1 μs. The entire system has been assembled and fired, but further work is needed on the diagnostics equipment.

Although preliminary, our work to date leads us to believe that the gas-embedded Z-pinch is feasible. Its basic simplicity makes the concept a potentially attractive approach to fusion if the instability problems can be controlled. We are thus maintaining the project at a modest level as part of our exploratory plasma research.

Key Words: Z pinch; gas-embedded Z-pinch; blast-containment reactor; 14-MeV neutron damage; liquid-lithium first wall; kink instability; sausage instability.

RISE: AN LLL CONCEPT FOR EXTRACTING OIL FROM OIL SHALE

We are working on an *in situ* process for recovering oil from thick deposits of moderately low-grade oil shale. The concept, called rubble *in situ* extraction (RISE), consists of breaking up the oil shale into rubble underground by mining methods, then retorting the rubble in place. With this process it should be possible to develop deposits having only 85 litres of oil per tonne of shale (20 gal/ton); commercial aboveground retorting processes typically require 125- to 145-litre/tonne (30- to 35-gal/ton) oil shale. Deposits 120 m or more thick containing oil shale of 85 or more litres per tonne in Colorado's Piceance Basin contain 50 × 10^9 tonnes (350 billion barrels) of recoverable oil. This is more than the recently quoted Middle East recoverable oil reserves of 45 × 10^9 tonnes (316 billion barrels).[5]

We have defined the technical and economic problems of oil-shale rubblization and retorting and have proposed a program to develop the RISE process for commercial application.[6] We estimate that this process could be developed in about 6 years at a cost of about $80 million with a high probability of success. Successful development would make possible the production of shale oil at lower economic and environmental costs than conventional surface retorting and would add significantly to our domestic oil supply.

The only practical method to obtain oil from oil shale is to heat it to the temperature (350-400°C) at which kerogen — the organic material in shale — decomposes to yield shale oil, gas, and residue. Because oil shale is a poor conductor and is essentially impermeable before retorting, it is necessary to provide heat-transfer surfaces and space for the circulation of a hot fluid. This can best be done by introducing both fractures and space into the oil shale — a process we call rubblization. Fractures alone provide a surface but no room to circulate a hot fluid.

For surface retorts, adequate rubblization is easily achieved by mining the shale and crushing it to a suitable size. *In situ* rubblization is more difficult technically but has potential economic and environmental advantages. The problem is to provide fragments of suitable size in a confined volume with enough space between them to allow for circulation of a hot fluid at modest pressure differences. If the fragments are too large, they will take too long to heat; if too many fine fragments are present, the pressure drop required for fluid circulation will be too large for a given void space. Void space must be adequate for circulation of fluids but should be kept low to minimize costs.

Various *in situ* processes have been proposed and studied for oil-shale recovery, among them fracturing of the rock with high explosives (conventional or nuclear) and/or massive hydraulic injection followed by combustion in place,[7] leaching methods, and the use of hot gases in permeable oil-shale regions.[8] Occidental Petroleum Corporation has reported field tests on a modified *in situ* process in which a portion of the oil shale is mined underground and the remainder blasted into the void so created, resulting in a rubble-filled column that is then retorted in place using combustion.[9]

We have coined the term "rubble *in situ* extraction" (RISE) for our version of this general procedure. With RISE, a rubble is created by continuous mining using a modified sublevel-caving technique (see frontispiece and Fig. 7). Approximately 20%, perhaps less, of the oil shale is removed to the surface, where it may be discarded directly or retorted in surface equipment and then discarded, depending on the economic and environmental conditions at the particular site. We believe that this technique can produce large volumes of rubble with uniform and controlled void space, suitable for commercial-scale retorting of thick resources — 120 to 600 m — in blocks about 100 m square. The rubble volume is then retorted in place using hot gas generated continuously by burning a portion of the oil shale in an air stream. Alternatively, an inert gas may be heated by external combustion, then circulated through the rubble.

A significant difference in RISE is the use of a continuous mining technique capable of preparing uniform rubble on a large scale, as opposed to mining out single or several larger cavities and blasting rubble into them. The latter procedure may be suitable for relatively thin, accessible beds of oil shale or for experimental field tests, but it is not expected to yield

Contact Albert J. Rothman (Ext. 8801) for further information on this article.

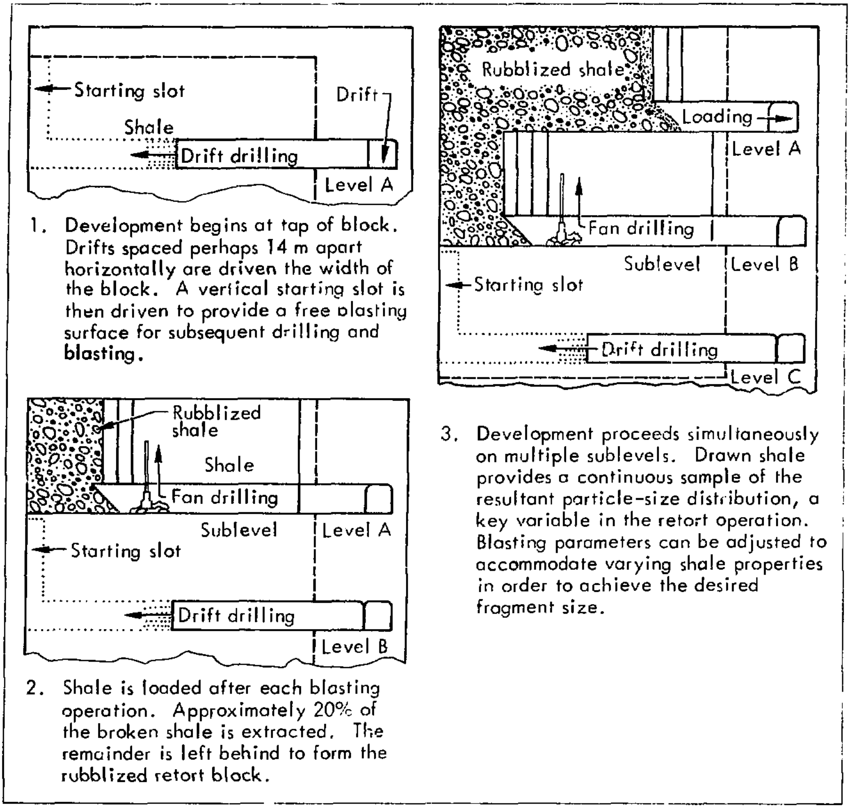


Fig. 7. Modified sublevel-caving mining method proposed for use in the RISE process.

the control or uniformity of the modified sublevel-caving technique, nor will it permit the continuous measurement of rubble size needed for adjusting explosives as mining proceeds.

Comparison with Other Processes

Tables 1 and 2 compare RISE with other processes for recovering oil from oil shale. The RISE process has many advantages. Nuclear fracturing is technologically promising but may require more detailed environmental study than nonnuclear processes.

We have chosen to concentrate on the RISE process for several reasons. 1) It utilizes a very large, thick resource compared with most other oil-shale approaches. Some 50 X 10^9 tonnes (350 billion barrels) of oil are recoverable using the technique, assuming a 70% resource recovery, 60% oil yield for *in situ* operations, and 95% oil yield for surface retorting of the mined shale. 2) The environmental impact of RISE is much smaller than for surface processes. Less shale needs to be dumped above ground, less water is required, and the population influx needed is significantly smaller. 3) Although

Table 1. Estimated costs for various oil-shale recovery methods

Method	Cost per tonne (per bbl)[a]	Capital investment for 14 000 tonnes (100 000 bbl) per day
Mining and surface retorting	$1.70 ($12)	$900-$1350 million
Rubblization by mining and *in situ* retorting (RISE method)	$1.15 ($8)	$540 million
Nuclear fracturing and *in situ* retorting	$0.70 ($5)	$270 million

[a]Cost includes upgrading to petroleum refinery feedstock standards.

costs are difficult to compare in times of rapid inflation, they appear to be significantly lower for RISE – $0.85 to $1.20/tonne ($6 to $9/bbl) – than for aboveground retorting processes. 4) There is reasonable likelihood of technological success, based on the experiments in the 135-tonne (150-ton) Laramie retort[10] and the encouraging reports on field tests by Occidental Petroleum Corporation.[9] Other *in situ* processes do not appear to offer this likelihood. 5) Capital, equipment, and large-scale mining capability are in short supply. The requirements for these are much lower for RISE than for surface processes. For these reasons, as well as the greater public acceptance of nonnuclear approaches, RISE offers the best likelihood of early commercial operation on a large scale. We estimate development to commercial-scale demonstration could be accomplished by 1980 or 1981.

The RISE concept and surface retorting processes may complement each other, in that the latter most effectively employ richer oil shale closer to the surface, while the former utilizes deeper, thicker deposits that are leaner in grade.

Technical and Economic Issues

We have identified a number of areas that need to be studied to develop and optimize the RISE concept and have developed an experimental and calculational approach to these issues. If the RISE technology is to be developed for commercial application, large-scale field tests will ultimately be required.

Because field experiments are costly, however, our approach is to develop our predictive computer model for the retorting process, do laboratory- and pilot-scale experimental work to confirm and improve the accuracy of the model, and use the model to guide future selection of field-test conditions and, ultimately, commercially viable operating conditions. Much of this experimental work has begun under the nuclear program, as reported in the following article.

Retorting. We think that the best method of retorting is to use air combustion of the residual coke

Table 2. Comparison of the major methods for recovering oil from oil shale. The analysis assumes a production rate of 140 000 tonnes (1 million barrels) of oil per day

Recovery method: Where retorted	Recovery method: Shale preparation	Oil shale resource: Grade, litre/tonne (gal/ton)	Oil shale resource: Oil, tonnes (10^9 bbl): In place	Oil shale resource: Oil, tonnes (10^9 bbl): Recoverable	Environmental impact: Annual disposal requirement, 10^9 tonnes (10^9 tons)	Environmental impact: Population influx	Environmental impact: Annual water use, 10^6 kg (acre-ft)
Surface	Underground mining, room and pillar	125 (30)	14 (100)	8 (54)	0.45 (0.5)	130 000	220 (176 000)
	Open-pit mining	85 (20)	100 (700)	94 (660)	1.1 (1.2)	140 000	215 (173 000)
In situ	Mining rubblization (RISE process)	85 (20)	100 (700)	50 (350)	0.21 (0.23)	100 000	120 (96 000)
	Nuclear fracturing	85 (20)	90+ (650+)	27+ (200+)	None	55 000	80 (65 000)

in oil shale to provide the hot gases to retort the shale. The proper operating conditions for commercial retorting need to be identified, including how much air, oxygen, and recycle gas is required per tonne of shale, what the oxygen content of the circulating air and the rate of retorting should be, and what is the optimum combustion temperature. The alternative retorting method – using externally heated hot gas – also needs study.

As the flame front moves down through the shale in a retorting column, the temperature in the shale as a function of vertical position changes accordingly. It is not known whether the temperature profile reaches a steady shape and continues that way down the column or flattens out below the flame front. In the latter case, the shale in the lower part of the chimney would heat up much more slowly than the rest. There is evidence that oil yield and quality may be affected by the rate of heating, so the temperature profile in the rubble column may have an important bearing on process economics.

Another problem area is that packed beds of oil-shale particles under high stresses may become compacted and lose permeability when heated. This could increase the pressure drop and decrease the gas flow rate through a thick rubble chimney, thus reducing the retorting rate. Pilot-scale studies of combustion and hot-gas retorting in packed rubble columns are now under way.

We have developed a computer model of the combustion-retorting of single blocks and of packed beds of blocks. This model will be improved and tested using existing experimental data and new data emerging from the pilot-scale retorting studies. Field data must then be obtained and used for further testing and refinement of the model.

Also, large volumes of stack gas will be discharged in retorting. Experimental studies indicate that the heating value of this gas may be less than 20 J/m^3 (50 Btu/ft^3). Increasing this value by modifying the retorting conditions or by reducing the nitrogen and carbon dioxide content of the retort gas would yield a more valuable fuel and thus reduce gas pumping costs.

Rubblization. A key problem in the *in situ* retorting of oil shale is to achieve a desired particle-size distribution combined with a porosity high enough to give a minimum pressure drop. Ideally, porosity should be evenly distributed to produce uniform burning and flow distribution over the retort area.

A totally *in situ* process requiring no underground mining does not appear technically feasible at this time (with the possible exception of nuclear rubblization). Void space must therefore be provided by mining a fraction of the shale. Permeability for retorting will be obtained as this void space is distributed in the rubble during the blasting and drawing process. Because a large-scale mining effort will be required, mining considerations such as ground control, subsidence or caving of the shale beds, ventilation, and safety must be addressed. Mining problems peculiar to *in situ* retorting, such as maintaining retort integrity, must also be given intensive study, observation, and analysis.

To obtain the necessary permeability, about 20% of the oil shale in the retort area will need to be extracted. Consequently, the degree and nature of subsidence must be examined to determine its effect on underground and surface structures, land forms, and groundwater aquifers.

Measurement and Control. Remote instrumentation is required for several purposes. After the rubble chimney has been formed underground, rubble size and void distribution need to be measured, at least in the development program. To follow the course of retorting and to provide data for a model, measurements of temperature, pressure, gas composition, and flame-front location are needed at various locations in the rubble chimney.

To make such measurements remotely in underground chimneys is not straightforward. Shifts in the rubble bed may damage or destroy *in situ* sensors, which will be difficult to replace. Thus, some effort is required to develop instrument systems suitable for *in situ* field measurements.

The experimental approach proposed for testing and improving our calculational retorting model is to measure the symmetry and velocity of the burn front and the composition of the gas at the burn front. The spatial location of the burn front could be measured with a three-dimensional array of thermocouples placed in the rubble. The limitations of temperature measurements using thermocouples need to be investigated, along with methods of processing the thermocouple signals. Other methods of measuring temperature must also be investigated.

The second part of evaluating the retorting model is to measure the gas composition at the burn front. Two steps are involved. First, to remove the gas sample from the region of the burn front, we currently envision using a gas-collecting bottle lowered into a perforated casing. At the desired sampling location, a remotely actuated valve would open to collect a sample to be brought to the surface. This scheme will require development work in the area of high-temperature seals and valves.

The second step is then to develop a method for analyzing the sample. Gas chromatography is one possibility, although mass spectroscopy may be desired.

Environmental Effects. Although *in situ* processing of oil shale is considered to have significant environmental advantages over mining and surface processing, studies will be needed to determine its environmental effects and to identify the steps required to minimize them. The cost of these steps must then be balanced against their anticipated benefits.

The major environmental issues to be addressed are the effect of the process on air and subsurface water quality, management of water consumption, disposal of any high-salinity ground water encountered, surface disposal of spent shale, leakage of exhaust gases underground, subsidence, and population influx.

Key Words: oil shales; oil shales – combustion; oil shales – heating; oil shales – pyrolysis; oil shales – retorting; oil recovery – nuclear methods.

PROGRESS IN LLL OIL-SHALE RESEARCH

The Laboratory's rubble *in situ* extraction (RISE) concept for recovering oil from oil shale (summarized in the preceding article) is supported by experimental work begun in 1973 as part of our nuclear rubblization program.[6]

We have proposed a kinetic model of kerogen decomposition that accounts for the observed thermal induction time of the kerogen. We are working on a computational model of the retorting process. To perform experimental studies on retorting, we have operated one pilot retort and are nearing completion of a larger retort. We have investigated in some detail the anomalously rapid heating observed in single oil-shale blocks. We are studying the pressure drop in rubble columns and have built equipment to examine the loss of permeability by oil shale under certain conditions.

Kinetics

A number of analyses of the kinetics of kerogen decomposition have been made[11-13] based on the data of Hubbard and Robinson's now classic experimental study.[11] These analyses, however, have not explicitly taken into account the thermal induction period -- the time it takes to heat up the sample in the furnace. Instead, several of them have implicitly assumed that this time lag arises from an autocatalytic mechanism. We have proposed a kinetic model, using two first-order equations, that accounts for the thermal induction time and thereby gives a simple yet accurate representation of the data.[14]

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Some typical results using our kinetic model are given in Fig. 8. These results demonstrate that our simple analysis fits the experimental data very well.

Retort Modeling

Numerical or computer models of oil-shale retorting have been developed by others, but a number of them are proprietary to various companies (and thus details about them are difficult to obtain) or are not considered workable.[15] Our modeling studies are a keystone in the RISE program. Th far, we have modeled the case of hot, inert gas heating a bed of uniform-size particles. In this model, hot gas enters the

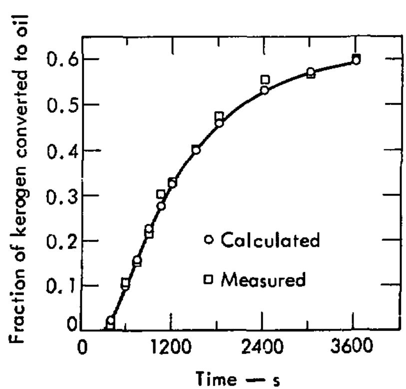

Fig. 8. Comparison of measured and calculated pyrolysis of oil shale at 425°C. The calculated values were obtained using our simple kinetic model.

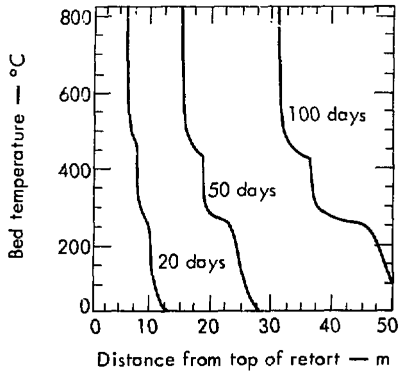

Fig. 9. Temperature profile as a function of time during retorting of rubblized oil shale in an *in situ* column for 0.2-m fragments, an input gas temperature of 827°C, and a gas input rate of 9 $g/m^2 \cdot s$ (1.4 $scf/ft^2 \cdot min$). The curves show the development of two major changes in slope as the temperature front moves down the column. The upper one, near 500°C, corresponds to the beginning of dolomite decomposition; above 500°C the decomposition is nearly complete. The lower break, near 300°C, corresponds to the decomposition to oil, gas, and char; above 350°C kerogen pyrolysis is virtually complete.

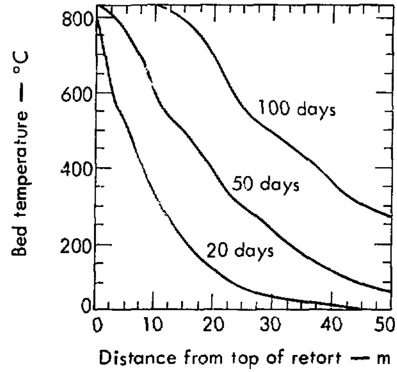

Fig. 10. Temperature profile as a function of time for the same conditions as in Fig. 9 except that the fragment size is 2.0 m. In this case the very large particles act as a thermal sink, causing extensive broadening of the temperature profiles.

retort at the top. The model describes the transfer of heat from the gas to the oil shale, the heating of the shale, the decomposition of the kerogen to gas and oil, the further heating of the shale, and the decomposition of mineral carbonates.

We have numerically solved the basic differential equations in the hot-gas retorting model; Figs. 9 and 10 show typical computer results. The temperatures plotted are averages of the outermost one-third of the particle volume. These figures show that extensive broadening of the temperature profiles occurs for very large particles. The figures are only representative of possible output from the retorting model. Still, this model allows us to study the effect of gas rate, input temperature, bed height, and particle size over broad ranges.

We have also developed a computer model of combustion retorting, in which the hot gas is produced by combustion in the bed. In this case, air entering the top of the bed burns the carbonaceous residue that remains after the kerogen has been retorted by the hot gas swept ahead of the burn front. Carbonate is decomposed in the same region in which the carbonaceous residue burns. At the leading (lower) edge of this combustion front, the remaining oxygen in the gas stream is consumed in burning some of the oil released in this region. Finally, in advance of (below) the combustion front, kerogen decomposes to oil, gas, and carbonaceous residue.

Both retorting models thus far apply to oil shale of uniform particle size. We are currently refining them to account for shale beds of mixed particle size.

Retorting Studies

Studies are under way at LLL to determine the effect on oil yield and product composition of different heating rates and atmospheres. This work is needed because there is a conflict in the literature and in unpublished data regarding the possibility that long-term heating of oil shale produces severe losses in oil yield.

In another area, the Bureau of Mines at Laramie, Wyoming, has reported a number of experiments using large retorts (9- and 135-tonne capacity)[10,16-18]; their results are very useful guides to retorting simulated rubble beds and have served as the basis for predicting yields in larger-scale field operations. However, a number of points need to be explored further, such as the effect of adiabatic operation. Heat losses to the atmosphere in the 135-tonne retort varied from 5% to 33%, and a number of runs showed unaccountable heat-balance discrepancies up to 37%. Furthermore, the percent loss increases with reduced retorting rates, because longer times are available for heat loss to take place *to the surroundings*.

To investigate these effects we have constructed a pilot retort 0.3 m in diameter by 1.5 m high and have made a number of exploratory runs. This 125-kg retort has a data-acquisition system and will be operated adiabatically as soon as the computer control system is completed. Some early results are given in Fig. 11 for an insulated column, but one whose heat losses to the atmosphere are appreciable. Thus, the temperature profiles will be different from those in a truly adiabatic column. Figure 11 shows an interesting feature: two humps or peaks in temperature occur when the gas produced is recycled to the top of the retort. We attribute the first peak to the combustion of components in the gas and the second to combustion of char lower in the retort.

Construction of a larger retort – 0.9 m in diameter by 6 m high – is nearing completion. Its purpose will be to permit insertion of larger blocks of rubble. We plan to instrument the interior of these blocks as well as the bed as a whole.

Single Blocks

Large single blocks of oil shale, when retorted in air, are heated much more rapidly than would be predicted by considering the heating mechanism to be one of simple conduction.[17,19] This more rapid heating may permit oil to be recovered efficiently from large blocks. Because of the potential importance of this behavior for *in situ* processing, we have investigated possible mechanisms for the temperature increase.

We have found that known mechanisms are unsatisfactory in explaining the observed behavior. For example, the transport of oxygen by diffusion from the block surface to the interior is too slow to produce the rapid temperature increase by oxidation of hydrocarbons. We therefore sought some rapidly varying or oscillatory phenomenon. We sealed pressure-sensing capillary tubes into a block of shale so that we could observe the interior pressure. During retorting of the block, pressure pulses occurred periodically. During part of each pulse, the interior pressure was lower than the pressure at the block surface. Because a shale block develops numerous small cracks during the early part of the retorting process, the pressure pulses result in transport of thermal energy and oxygen to the interior of the block along these cracks. We have observed this pulsing behavior in a range of block sizes and shale grades. Its amplitude is strongly dependent on the richness of the oil shale and on the size of the oil-shale blocks. Pulses are strongest for the richer and larger blocks.[20]

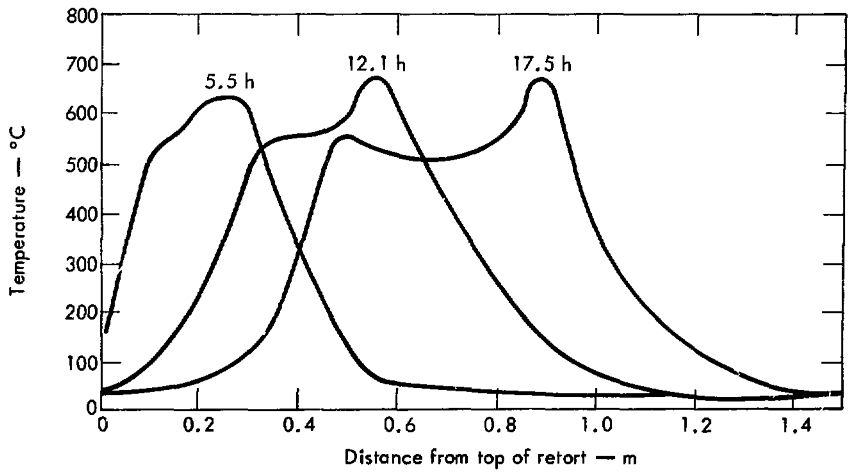

Fig. 11. Temperature profiles in the LLL 125-kg retort at various times after ignition. The temperatures are measured in a thermowell; the actual temperatures are about 150 to 200°C higher. The first peak is due to the combustion of components in the gas; the second represents the combustion of char lower in the retort.

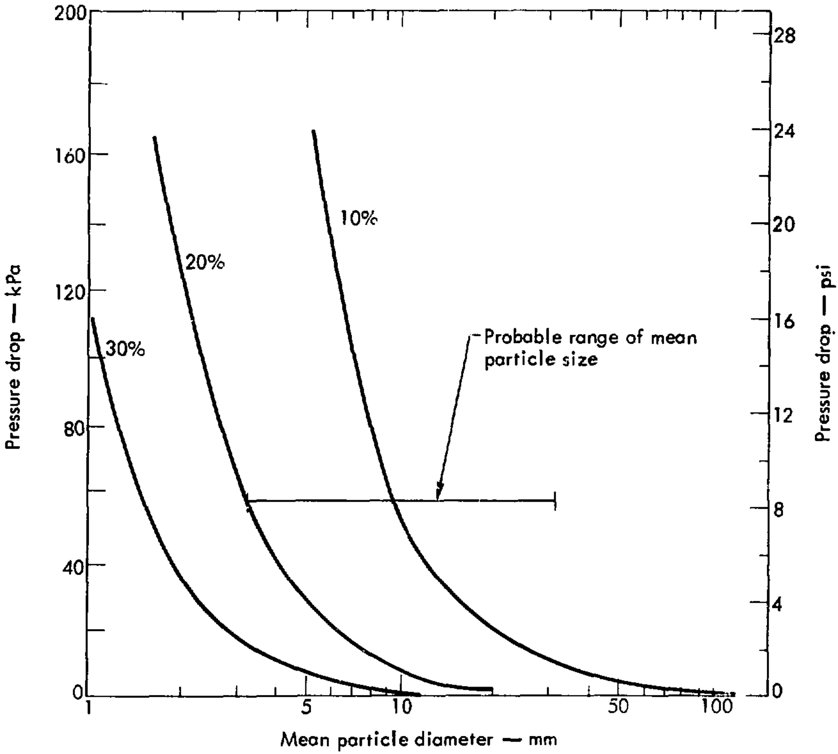

Fig. 12. Calculated pressure drop as a function of mean particle diameter for retorting of oil shale in a 300-m-high retort, assuming 500 m^3/tonne (16 000 scf/ton) of air plus recycle gas and an air velocity of 7.5 mm/s (89 ft/h). This corresponds to a 60-75 cm/day retorting rate for a 10 to 30% porosity. Also, we assume that the flow rate of air plus recycle gas is proportional to the retorting rate and that the pressure drop is approximately proportional to the flow rate of air plus recycle gas. Also shown is the probable range of mean particle sizes in an *in situ* retort.

We have used the results of these experiments in our model of the combustion-retorting process.

Pressure Drop in Rubble

Although not strictly an experimental program so far, this pressure-drop study has been important in determining the drop to be expected in rubble columns. A key issue is how to characterize the mean particle size of a bed having a broad particle-size distribution. We have found[21] that the Ergun equation[22] will adequately predict the pressure drop as a function of flow rate provided that the mean particle diameter (d_m) is taken as the harmonic or surface-area mean:

$$1/d_m = \sum_i (W_i/d_i),$$

where W_i is the weight fraction of the particles having diameter d_i.

Figure 12 shows the calculated pressure drop as a function of particle size for various oil-shale porosities.

The probable range of mean particle size is also shown, based on estimates of particle sizes of mine-run shale and of hard rock subjected to an underground nuclear explosion.[21] Neither situation is truly applicable, but they give some idea of the range to be expected. These pressure drops were calculated for typical gas flow rates, based in part on pilot runs in the Laramie 135-tonne retort.[10]

We have also calculated curves showing the electrical cost for pumping as a function of the unit cost of electricity and the pressure drop in the rubble column. The cost penalties for high pressure drops are significant.

Deformation and Loss of Permeability

From Bureau of Mines work,[23,24] it appears that the permeability of 130-litre/tonne (31-gal/ton) and richer oil-shale rubble beds decreases drastically when the shale is heated at a rate of about 70°C/h under a stress on the order of 1 MPa (hundreds of psi). Measurable but much smaller losses in permeability are noted in 85-litre/tonne (20-gal/ton) oil shale under the same conditions. There is also evidence of some recovery of permeability with prolonged heating.

We plan to study this phenomenon in some detail, including the effect of slower heating rates that correspond to those expected in an underground, *in situ* case. Our equipment has been constructed and experiments have started.

Key Words: oil shales; oil shales – combustion; oil shales – heating; oil shales – pyrolysis; oil shales – retorting.

RADIONUCLIDE ACCUMULATION IN OYSTERS

One thrust of our Biomedical and Environmental Research Program is to pursue studies into possible effects of energy-related pollutants on the marine environment. Industrialization and urbanization have, for example, increased the amounts of trace-metal pollutants released into estuarine environments such as bays and submerged river valleys. Such pollutants are quite capable of upsetting the delicate balance of the marine ecosystem. Our concerns have been twofold: effects on the marine organisms themselves and, through the food chain, effects on man.

Reported here are results from a study of the accumulation of radioactive trace metals by oysters from soluble and particulate materials in seawater. Oysters are effective concentrators of many trace metals. Measurement of trace contaminants is simplified when the contaminents are radioactive. For our experimental site we chose the discharge canal of Pacific Gas and Electric Company's boiling-water-reactor power plant at Humboldt Bay, California. Monitored in the study were concentrations of four radionuclides that differ in their physicochemical properties: manganese-54, cobalt-60, zinc-65, and cesium-137.

Our results indicate that suspended particulates play an important role in the accumulation of some radionuclides and that resuspension of particles from bottom sediments is an important source of radionuclides between release periods. Any model of the uptake by marine organisms of radionuclide or trace-elements that have a high affinity for particulates will have to take into account the sediments as well as the water mass.[25]

Oysters are filter-feeding animals and, as such, maintain a steady flow of water through their gills for feeding, respiration, and removal of metabolic byproducts. The water transport rate through the gills of an adult oyster may vary from several litres per hour up to about 34 litres per hour.[26] Pollutants enter these animals by ingestion of living and nonliving particulates in suspension in seawater and by sorption of substances dissolved in the water. This sorption occurs directly by the accumulation of ions or small molecules by exposed tissues or indirectly by the adsorption of soluble material onto the mucous sheets on the surface of the gills with the subsequent ingestion of these sheets.[26]

Our experimental site, the Humboldt Bay Power Plant, is located about 8 km southwest of Eureka, California, on the east shore of Humboldt Bay (see Fig. 13). The plant produces electricity with two 54-MW_e fossil-fuel units and a single 65-MW_e boiling-water reactor. Cooling water from the south part of the bay is pumped from an inlet canal through the condensers of all three generating units and then discharged into a short canal leading back to the central part of the bay (see Fig. 13).

Radioactive wastes from the stack, the power building, the refueling area, and laundry operations are accumulated and processed at the plant. Batches of low-level liquid waste are released into the discharge canal waters at irregular intervals. The radiation levels at release are in accordance with limits prescribed by ERDA and by the California North Coastal Water Control Board.

The discharge canal is 140 m long and 12 to 20 m wide depending on the tides. Its bottom and sides consist of mud with a high content of organic material. The flow rate in the canal varies from 3000 to 7000 litres/s. The temperature of the water is generally about 22°C, but it can reach about 28°C at the experimental station when the effluent is heated as part of an operation to remove mussels from condenser lines.

The Experiment

The experimental station we installed in the discharge canal consisted of a redwood raft and an instrument shed. The raft (Fig. 14) had openings for suspending containers to hold the oysters in the water stream and to collect samples of settled particulates. All construction below the water line was nonmetallic to minimize contamination by trace metals.

The oysters we used were 3-year-old *Crassostrea gigas* obtained from commercial beds in the north part of the bay. We introduced several hundred of them into the discharge canal: half were placed in the water stream in plastic cages suspended from the raft and the other half in aquaria on the raft. The aquaria were

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supplied with water pumped through 1-μm filter cartridges at a rate of about 30 litres/min during the radioactive waste release.

We put the oysters in the canal 12 to 14 hours before a scheduled release to allow enough time for the animals to acclimate to the new environment. The animals were sampled immediately before and after the release and at 1-day intervals afterward. For each sampling we used the soft tissues from 50 to 75 animals.

Throughout the experimental period, we collected the suspended particulates on 1-μm filters. Settled

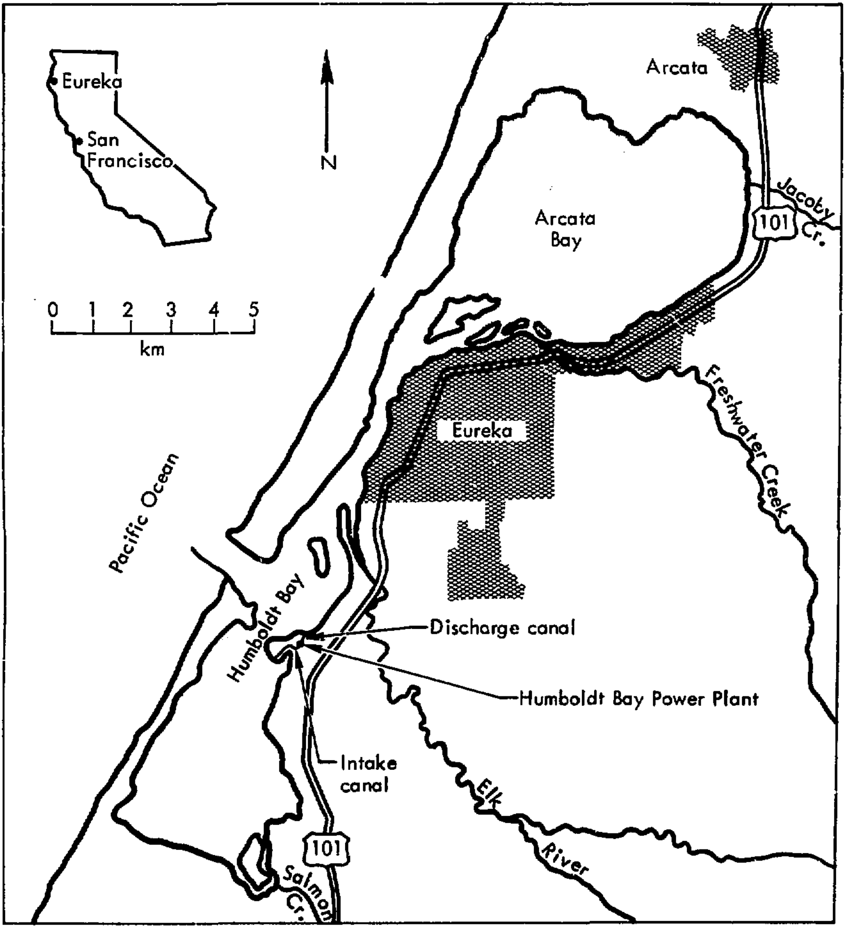

Fig. 13. Map of Humboldt Bay area of California, showing the location of the PG&E power plant and its discharge canal, in which our experimental station was located.

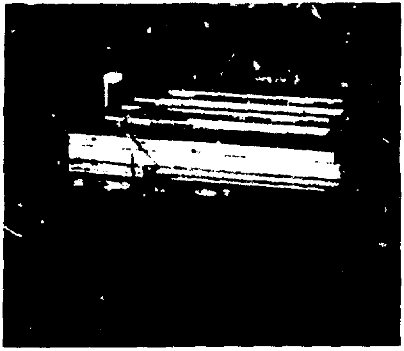

Fig. 14. Raft in discharge canal for oyster experiments. The raft, which is about 3 × 4 m, was held in place in the center of the canal by a system of lines and pulleys by which it could be pulled to shore at will.

particulates were taken from collection trays suspended from the raft at the same level as the animals in the baskets and from trays placed on the bottom of the canal. We collected water samples before, during, and after the release of radioactive waste. For each collection we filtered about 200 litres of seawater through 1-μm cartridges. The radionuclides in the particulates and the filtrate were analyzed separately.

All samples were counted on Ge(Li) detectors; chemical yields of cobalt, cesium, manganese, and zinc were determined by atomic absorption spectrometry.

Results and Discussion

The availability of some radionuclides to filter-feeding organisms appears to be determined by the amount of particulates in the water – living microorganisms, organic detritus, and inorganic material. We observed that for oysters in July, a period of high biological productivity, filtering the water decreased the quantities of radionuclides accumulated. During this month, in postrelease oysters, the concentrations in nonfiltered seawater were higher than in filtered seawater for all radionuclides (see Table 3). The accumulation rates for animals in filtered vs nonfiltered seawater were lower by about 65% for manganese-54, 90% for cobalt-60, 40% for zinc-65, and 30% for cesium-137.

Two sources of suspended material were available to the oysters in the discharge canal: particulates carried with the inflowing water and those resuspended from the material deposited onto the bottom. The particulate load of the inflowing water probably depends on the biological and physical processes

Table 3. Normalized radionuclide concentrations[a] in oysters maintained in the discharge canal of the Humboldt Bay Power Plant

		^{54}Mn	^{60}Co	^{65}Zn	^{137}Cs
July 31, 1973	Nonfiltered seawater				
	Prerelease (pCi/kg)	ND	ND	17	ND
	Postrelease (pCi/kg)	28	20	83	0.76
	Rate (pCi/h)[b]	14	10	33	0.38
	Filtered seawater				
	Prerelease (pCi/kg)	9.5	ND	22	ND
	Postrelease (pCi/kg)	19.4	2.6	61	0.52
	Rate (pCi/h)[b]	5	1.3	20	0.26
December 4, 1973	Nonfiltered seawater				
	Prerelease (pCi/kg)	11	3.3	40	ND
	Postrelease (pCi/kg)	9	4.3	67	1.1
	Rate (pCi/h)[b]	–	0.3	9	0.37
	Filtered seawater				
	Prerelease (pCi/kg)	13	ND	22	ND
	Postrelease (pCi/kg)	25	ND	67	1.1
	Rate (pCi/h)[b]	4	–	15	0.37

[a]Measured concentration (pCi/kg) divided by expected water concentration in discharge canal (pCi/litre). ND means no concentration detected.

[b]Amount accumulated per hour during the release.

occurring in South Humboldt Bay. Resuspension of bottom sediments appears to be related to changing hydrological conditions in the bay and the discharge canal.

In the suspended particulate fraction obtained from the water, comparisons of the observed radionuclide concentrations with those expected (based on the amount of each radionuclide released) indicate that the radionuclides differed in their affinity for the particulates. These differences in affinity are related to the chemical properties of the radionuclides and of the particulates. The differences in the observed percentages among radionuclides are similar to the differences in the water-sediment distribution coefficients determined for the radionuclides.[27]

The bottom sediments in the discharge canal represent a pool of radionuclides bound to particulates; this pool appears to control the concentrations of radionuclides in the water between releases. The availability of nuclides in this sediment to filter-feeding organisms is affected by a number of physical and chemical factors.[28] In the discharge canal, the important physical factors are the flow rate of water into the canal, the tidal level of the water in the bay, and the reworking of the sediments by the larger, indigenous, bottom-dwelling organisms. Important chemical factors are the specific chemical properties of the radionuclides and the metabolic activities of microorganisms in the sediment layer.

We observed that radionuclide concentrations in bottom sediments changed with time after a release, probably due in part to dilution with nonradioactive particles in the water flowing into the discharge canal. Over the same time interval we had measured the particulate loads in the intake and in the discharge canal water and found that the quantities of particulates in the intake were about the same as those in the discharge canal. These data indicate that the intake canal water contained enough nonradioactive particulates to reduce the concentration of radioactive particulates in the discharge canal when the two are mixed.

The radionuclide concentrations in the particulates in suspension and in the particulates settled onto the raft collection trays also varied with time – probably due to differing rates of resuspension of bottom sediments and to differing amounts of bound radionuclides in the sediments. Sediments are considered to consist of a historical (unmixed) layer and a mixed layer.[28] In the discharge canal the homogeneity of the mixed layer depends not only on physical and chemical processes but also on the frequencies of the radioactive releases and the magnitude of the differences in quantities discharged from release to release. If the bottom sediments in the mixed layer are heterogeneous and the depth of scouring of the bottom sediments varies with the tidal cycle, variations in concentrations like those observed would not be unexpected for the particulates deposited on our raft collection trays and filter cartridges.

We observed differences between July and December in the amounts of radionuclides accumulated in the oysters in the nonfiltered water (see Table 3); this was probably due to differences in both the quantity and composition of the particulates in the water. Both parameters are related to events occurring in South Humboldt Bay. Because the South Bay is shallow, light can easily penetrate to the bottom; the mud flats support extensive beds of eelgrass. Measurements of phytoplankton productivity and the standing stock of eelgrass during the previous year indicate that biological activity was higher in July and August than in December.[29] Increases in phytoplankton productivity would result in direct increases in the amount of suspended particulates; increases in eelgrass productivity would result in indirect increases (suspended detritus from the decomposition of decaying plant material).

It has been established for many bivalve mollusks that the presence of food material in the water results in circulation of water for feeding as well as for respiratory purposes.[26,30] Because primary productivity in Humboldt Bay is considerably greater in July than in December, it is very probable that in July more of the oysters in the discharge canal would be circulating seawater for feeding purposes. Consequently, the differences in accumulation between the July and December animals are probably related to the presence of the food material in the water.

The role of particulates in radionuclide accumulation differs with radionuclide. In oysters, cobalt-60 appears to be accumulated primarily from the suspended particulate fraction, whereas radiocesium appears to be accumulated primarily from the soluble fraction. Because the particulates vary in amount and composition both temporally and spatially in an environment, the quantities accumulated by organisms will vary correspondingly for those radionuclides with high affinities for particulates.

The turnover rates of radionuclides in bivalve mollusks have been measured in both the field and the laboratory by many researchers. Our results indicate that the rates obtained in the field will vary with the season. Rates determined in the laboratory would be expected to vary with the kinds and amounts of particulates available for food. More information is needed on turnover rates under varying environmental conditions before the proper rates can be selected for biological modeling.

Most models of the effects of pollutants released into ecosystems have considered the transport and movement of materials in the water mass. Our study shows that, for radioactive and trace-element pollutants that have a high affinity for particulates, modeling of both the water mass and the sediments may be more appropriate

Key Words: mollusks - oysters; mollusks - radioisotope uptake; Humboldt Bay Power Plant; nuclear power plants - environmental studies; radioisotope uptake; radioisotopes - contamination; radioisotopes - sorption; water pollution.

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